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DEFENCE RESEARCH ESTABLISHMENT OTTAWA
TECHNICAL NOTE 82-35

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ABSTRACT

The theory of the mobility of ions in gases is reviewed, and it is seen that the mobilities of large ions in air are beyond today's state of development of the theory. For large ions the concept of reduced mobility is questioned, and it is indicated that the customary semi-logarithmic presentation of experimental data is not theoretically justified.

RÉSUMÉ

La théorie de la mobilité des ions dans les gaz est revue, et il appert que la mobilité des gros ions dans l'air ne peut être expliquée par la théorie telle qu'elle existe actuellement. On s'interroage sur le concept de mobilité réduite des gros ions et on montre que la présentation semi-logarithmique habituelle des données expérimentales n'est théoriquement pas justifiable.

INTRODUCTION

The last few years have seen the development of automatic alarms for chemical warfare agents which detect these compounds by responding to ions of particular mobilities. Development has proceeded heuristically by simply measuring the mobility of the ion in question and programming the microprocessor of the alarm to respond to the appropriate peak. There is, naturally, some curiosity as to why these ions have the mobilities they do, and the aim of this note is to attempt to reconcile modern theory and the observed mobilities.

BASIC THEORY

An ion in a constant electric field is accelerated continuously. If other particles are present collisions will occur; in these collisions some of any ions' momentum and energy are transferred to the other particle. In this case the ions' instantaneous velocities are definitely not constant - they are accelerating between collisions and then losing velocity at each collision. Not very many collisions are required before the ions are travelling at a constant average velocity, which is called their drift velocity, v_d. By definition, their mobility, K, is their drift velocity divided by the electric field, E, thus

$$v_d = K E \quad . \tag{1}$$

Already we see that it is the details of the collisions which determine the drift velocity, and thus the mobility. This is an ominous note for the success of the theory we need, since collisions of complex molecules cannot be readily modelled. However there are a number of significant simplifications from the general situation described above which have been seen to apply to chemical agent alarms:

1. The number of ions is much less than the number of neutrals, i.e. non-charged particles. Thus we can ignore ion-ion collisions, which cuts the workload in half.
2. The drift velocity is much less than the mean thermal speed of the particles. In other words, the average speeds of the particles before and after collisions differ only negligibly from their thermal speeds. This greatly simplifies the mathematical considerations, as the following arguments show.

In this low-field case the mobility is independant of field strength, that is, the drift velocities are proportional to field strength. This can be seen by imagining that the field intensity is doubled. The ions then accelerate twice as fast. However they leave each collision with, on average, the thermal velocity. Thus their average incremental velocity (incremental above the thermal velocity) is doubled, and this incremental velocity is nothing but the drift velocity.

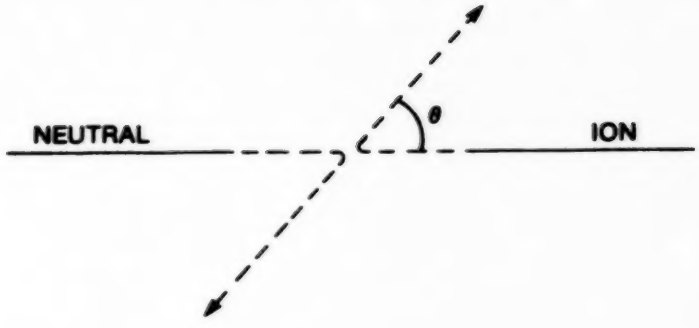

Figure 1. Collision between an ion and a neutral molecule in the centre-of-mass reference frame.

Also, in the low-field case the mobility is inversely proportional to the number density of neutrals. This follows because the drift velocity is proportional to the time between collisions, since after each collision the ion "starts again" from the thermal velocity. The time between collisions is inversely proportional to the number density.

The fact that the fields are low permits an easy approach to the consideration of collisions. The concept is that even though the ions are being accelerated by the field, they do not accelerate overall, since all the momentum that they gain from the field between collisions is transferred to the neutrals. It is convenient to consider these binary collisions in the centre-of-mass frame, illustrated in Figure 1. In this reference frame the particles approach or recede from the centre at velocities such that there is no net momentum. Thus if the ion, mass m_i, has velocity v_i and the neutral, mass m_n, has velocity v_n, then (v_r is the relative velocity)

$$v_i - v_n = v_r \qquad (2)$$

$$m_i v_i + m_n v_n = 0 \quad . \qquad (3)$$

After the collision the ion and neutral are moving at some angle θ from their original centre-of-mass directions. By considering the one-body problem* which is dynamically equivalent to this two-body problem (Ref 1, p 64) it can be shown that the momentum change of the neutral molecule (or of

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* This equivalence, and thus Equations 4 and 5, holds only for elastic collisions. The vast majority of collisions, even in atmospheric-pressure chemical ionization, are elastic.

the ion) in the direction that the ion was travelling is, in any collision

$$\Delta p = \mu v_r (1-\cos\theta) \qquad (4)$$

where μ is the reduced mass, $1/\mu = 1/m_i + 1/m_n$. If we now average over many collisions all the random components average to zero and we are left with the momentum in the direction of the field transferred to a neutral in an average collision

$$\Delta p = \mu v_d (1-\cos\theta) \qquad . \qquad (5)$$

(In this argument we are being rather careless about details of averaging and will continue in this vein to avoid obscuring the development.) The angle θ is determined by the impact parameter, b, (which is the perpendicular distance between the initial trajectories of the particles in the centre-of-mass frame) in a fashion determined by the details of the collision, and this relationship is complex to derive. We can say, however, that the average number of collisions one ion makes in a period t with impact parameter in the range b to b + db is $tNv_r 2\pi b\, db$ (N is the number density of neutrals), so the momentum that ion transfers to the neutrals is

$$2\pi N v_r \mu v_d t \int_0^\infty (1-\cos\theta)\, b\, db \qquad .$$

The momentum that that ion , charge e, receives from the field, E , in the same time t is eEt. Thus since all this momentum is transferred to the neutral particles

$$2\pi N v_r \mu v_d \int_0^\infty (1-\cos\theta)\, b\, db = eE \qquad . \qquad (6)$$

If we now define the collision cross-section, Q, as

$$Q = 2\pi \int_0^\infty (1-\cos\theta)\, b\, db \qquad (7)$$

and since, in the low-field case ($\bar{v}_r$ is the average relative velocity of all ion-molecule pairs)

$$\mu \bar{v}_r^2 = 3kT \qquad (8)$$

where k is Boltzmann's constant and T the temperature, Equation 6 gives

$$K = \frac{e}{\sqrt{3\mu kT}\, NQ} \qquad . \qquad (9)$$

This argument is correct in approach, but because the averaging procedures were not rigorous the correct coefficient is actually (Ref. 2, p 121)

$$K = \frac{3\sqrt{2\pi}}{16} \frac{e}{\sqrt{\mu kT}\, NQ} \quad . \tag{10}$$

This result appears simple, but only because all the complexity is contained in the cross-section, Q. The next section discusses calculation of cross-sections.

To conclude this section we will consider ions drifting in a mixture of gases, for example in air. From Equations 6 and 7 we can write, for the mobility of an ion in a mixture of gases

$$v_d \sqrt{3kT} \sum_j \sqrt{\mu_j}\, N_j\, Q_j = eE \tag{11}$$

where the index j runs over the number of gaseous components. For each component we can say

$$v_{d(j)} \sqrt{3kT}\, \sqrt{\mu_j}\, N\, Q_j = eE \tag{12}$$

where $v_{d(j)}$ is the drift velocity in pure component j whose number density has been increased to N, the total number density in the mixture. Thus for each component

$$\sqrt{\mu_j}\, Q_j = \frac{eE}{\sqrt{3KT}\, v_{d(j)}\, N} \quad . \tag{13}$$

Substituting Equation 13 into Equation 11 gives

$$\frac{1}{K_{mix}} = \sum_j \frac{X_j}{K_j} \tag{14}$$

where K_{mix} is the mobility of the ion in the mixture, X_j is the mole fraction of the j^{th} component, and K_j is the mobility the ion would have in pure component j at the full pressure. Equation 14 is known as Blanc's Law.

General references used in the preparation of this section and the next were the books by McDaniel (1) and McDaniel and Mason (2) for collision theory and an introduction to cross-sections, and a text on electrodynamics such as Jackson's (3) for multipole moments and interaction potentials.

THE COLLISION CROSS-SECTION

A collision between an ion and a molecule may be said to occur if the force they exert on each other is sufficient to change either of their velocities significantly. Clearly we need to consider the forces between these particles. They are totally electrical in nature, and can be classed in two groups, electrostatic and polarization.

1. Electrostatic: If both particles were charged then the force they exert on each other would vary as r^{-2} (r is their separation), and therefore the interaction potential as r^{-1}. This term is, of course, absent here since we are considering ion-neutral interactions. However one or both particles could have permanent dipole, quadrupole, or higher moments, and these charge distributions could interact with each other and with the ionic charge. These interaction potentials vary more rapidly with r, such as r^{-3} for the charge-dipole potential. Furthermore, the potential depends on the relative orientation of the particles, which makes it difficult to treat theoretically.

2. Polarization: A polarization force will arise if the ion, due to its proximity, can distort the electronic cloud of the neutral particle. If it is a dipole moment which is induced the resulting potential, called the induced dipole potential, varies as r^{-4}. This is the longest range of the possible polarization forces, but others occur, such as charge-induced quadrupole or higher moments, and permanent dipole or other moments on either particle inducing moments on the other. For example the dipole-induced dipole potential varies as r^{-6}, which is the same variation as the charge-induced quadrupole potential, r^{-6}. Polarization forces are always attractive.

There have been no attempts to calculate theoretical cross-sections using all of the possible forces as, unfortunately, is required for the drift of nerve agent ions in air. In fact virtually no calculations including the electrostatic forces have been done. It is informative, regardless, to review very briefly some of the calculations which have been done. The simplest case of all is for collisions in which the charge-induced dipole force is the only significant force This simple case leads to the so-called "polarization limit". Then we will move on to review briefly some of the approximate calculations which have been carried out.

If both particles are spherically symmetric there is no possibility of permanent moments of charge distribution, thus no electrostatic forces. If in addition temperatures are low then the energies involved in collisions are low, and then the longest range force predominates, which is the induced dipole force (the ionic charge induces a dipole moment in the neutral). The potential energy of this force is

$$V_p = -\frac{e^2\alpha}{2r^4} \qquad (15)$$

where e is the electronic charge and α the polarizability of the neutral. The calculation of the cross-section, Q, proceeds according to Equation 7, but is tedious to do because the angle of deviation, θ, must first be calculated as a function of the impact parameter, b. The result (Ref 2, p 146) is

$$Q_p = 1.0711 \frac{e}{N} \sqrt{\frac{\alpha}{kT}} \tag{16}$$

which gives for the mobility

$$K_p = 0.4388 \sqrt{\alpha\mu} \tag{17}$$

for α in nm^3 , μ in g/mole, and K_p in $cm^2\ V^{-1}\ s^{-1}$. This value is called the polarization limit mobility for ions of reduced mass μ in a gas of polarizability α . The use of the term "limit" is imprecise here, though, since in more complex cases the mobility can be greater or smaller than this value. However the polarization-limit value can be useful as a first estimate, and for comparisions of experimental mobilities the ratio K/K_p can serve as a useful "normalized mobility".

Even in this simplest case the calculation of the cross-section, Q, is too lengthy to present, and the calculation for a case such as the 12-6-4 potential,

$$V = \frac{\varepsilon}{2}\left[(1+\gamma)\left(\frac{r_m}{r}\right)^{12} - 4\gamma\left(\frac{r_m}{r}\right)^{6} - 3(1-\gamma)\left(\frac{r_m}{r}\right)^{4}\right] \tag{18}$$

which is known to represent accurately the potential in some situations, is significantly more complicated (ε and r_m are the depth and position of the potential minimum and γ is a dimensionless parameter). A precise test of any calculated mobility is to measure the mobility over a range of temperatures since increasing the temperature means that the shorter-range forces become more important (Ref. 4,5,6). It seems fair to say that, even with three adjustable parameters, the resulting cross-section does not agree well with experimental mobilities over a range of temperature (Ref. 7). A further complexity which some authors (Ref. 8) have considered is the existence of a bulky core as might be found in polyatomic ions. The expression

$$V = \frac{\varepsilon}{2}\left[\left(\frac{r_m - a}{r - a}\right)^{12} - 3\left(\frac{r_m - a}{r - a}\right)^{4}\right] \tag{19}$$

in which a is the diameter of the rigid core, describes the potential energy of these cases quite accurately. Again, however, there are three adjustable parameters, and again agreement is not satisfactory (Ref. 7). There are at least two other significant factors which produce still further complexities (Ref. 9) - orientation-dependant terms and electrostatic potentials. Few, if any, studies incorporating these aspects have been done.

This very brief review of collisions only slightly more complicated than the induced-dipole case, with comparision with recent experimental data, shows that only the simplest cases can be handled accurately by modern theoretical techniques.

ACCURACY OF REDUCED MOBILITIES

It is usual, in mobility studies, to tabulate so-called reduced mobilities, which are mobility data reduced to standard temperature and pressure to provide a common basis for comparision. This is done by application of the ideal gas law,

$$K_o = K \frac{P}{760} \frac{273}{T} \qquad (20)$$

where K_o is the reduced mobility and P and T are the pressure and temperature at which the mobility K was measured. The mobilities published in this note are reduced mobilities.

This correction to standard conditions corrects only for the number density of the gas molecules. However we have already noted that, since the temperature controls the distance of closest approach during a collision, it affects the scattering according to the variation of potential with distance. Experimental results have been mentioned which disagree with even sophisticated theories of mobility variation with temperature, and definitely disagree with Equation 20 above.

The concept of reduced mobility, we conclude, should be applied only to ion-neutral combinations which are well described by the polarization limit (or other temperature-independant mobilities). For complex ions or gases the variation of mobility with temperature is more complex.

APPLICATION OF MODERN THEORY TO IONIZATION DETECTION OF CHEMICAL WARFARE AGENTS

While the theory discussed above can be applied to any ion in any gas, the cross-section, Q, can be calculated accurately for only the simplest cases. There are at least four reasons why these calculations would be very inaccurate for nerve agent ions (or those of other chemical warfare agents) in air:

1. Both nitrogen and oxygen have quadrupole moments, while the ion

has a dipole moment. These electrostatic forces will be at least as large as the induced dipole force. Revercomb and Mason (10) give an example of a spherical ion, K^+, whose mobility is reduced 28% in nitrogen by the quadrupolar interaction.

2. Neither the ion nor the molecule are spherical, so orientation effects will be significant.
3. Agent ions have a bulky core (11).
4. Some of these ions undergo reactions while drifting, which results in an average mobility of the species involved. This aspect introduces several other complexities, such as how the kinetics are affected by temperature, and the effects of the concentrations of other components.

Thus we conclude that the only role of the theory presented above, in design of detectors for chemical warfare, is to provide a physical picture of ion mobility in gases. The collisions of complex ions with diatomic molecules is far beyond modern theory. Development of ionization-type detectors must rely on experimentally measured mobilities. Furthermore, these mobilities must be measured near, or preferably throughout, the temperature range in which the detector is expected to operate.

EMPIRICAL RELATIONSHIPS BETWEEN ION MOBILITIES AND MASSES

Ubiquitous amoung scientists and engineers is the desire to establish tidy correlations in experimental data. Measured ion mobilities are treated no differently. Unfortunately, as we have seen, there is no precise theoretical relationship between mobility and ion mass. Nevertheless, various empirical relationships are used, and in this section we will leave behind the precise theory to examine them.

Speaking generally, large ions have large collision cross-sections and thus low mobilities, and large ions are heavy ions. Thus some correlation between ion mobility and mass must exist, even though the details of the relationship are to be found only in the collision calculations. It has become an established custom, at least in agent ion studies, to plot mobility against the logarithm of the ion mass. Data for a number of ions* which occur in studies of nerve agent ions, mustard ions, and ions of a simulant (Ref. 12,13) are plotted against the logarithm of the ion mass in Figure 2, and tabulated in Table 1. The ion masses were determined by a mass spectrometer coupled to the drift tube. While the graph does show reasonable correlation, this particular functional relationship is not the only one to do so. Correlation is also shown with

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* All but one of the mobilities listed in this note are from undistributed reports (Ref. 12) produced by PCP Inc. under contract to the British Government. The data on VX is from Ref. 13.

TABLE 1

Mobilities in Air of Ions Found in Agent and Simulant Studies

Molecule	Ion[2]	Mass (g/mole)	Reduced Mobility ($cm^2 V^{-1} s^{-1}$)
GB	M_3H^+	421	1.04
	M_2H^+	281	1.35
	$M(Ac)H^+$	199	1.57
	$M(W)H^+$	159	1.72
GD	M_2H^+	365	1.11
	$M(Ac)H^+$	241	1.38
GA	M_2H^+	325	1.23
	$M(Ac)H^+$	221	1.46
	$M(W)H^+$	181	1.62
TEP[1]	MH^+	365	1.14
	$M(Ac)H^+$ [3]	241	1.45
	MH^+	183	1.68
VX	MH^+	268	1.30 [4]
MS[1]	MO_2^-	184	1.60
H	MO_2^-	191	1.59

(1) TEP = Triethyl Phosphonate, MS = Methyl Salicylate
(2) M = parent molecule, W = water, Ac = acetone
(3) not mass-identified
(4) recorded at 150°C, all others at 40°C. See text for discussion of temperature variations.

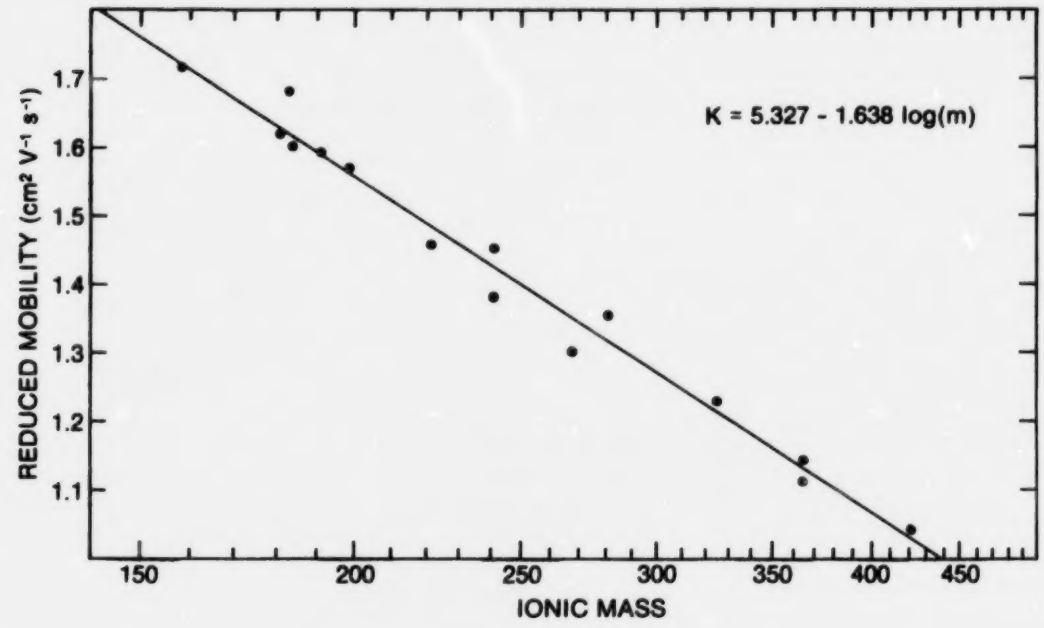

Figure 2. The reduced mobility of agent and simulant ions plotted against their masses on a logarithmic scale.

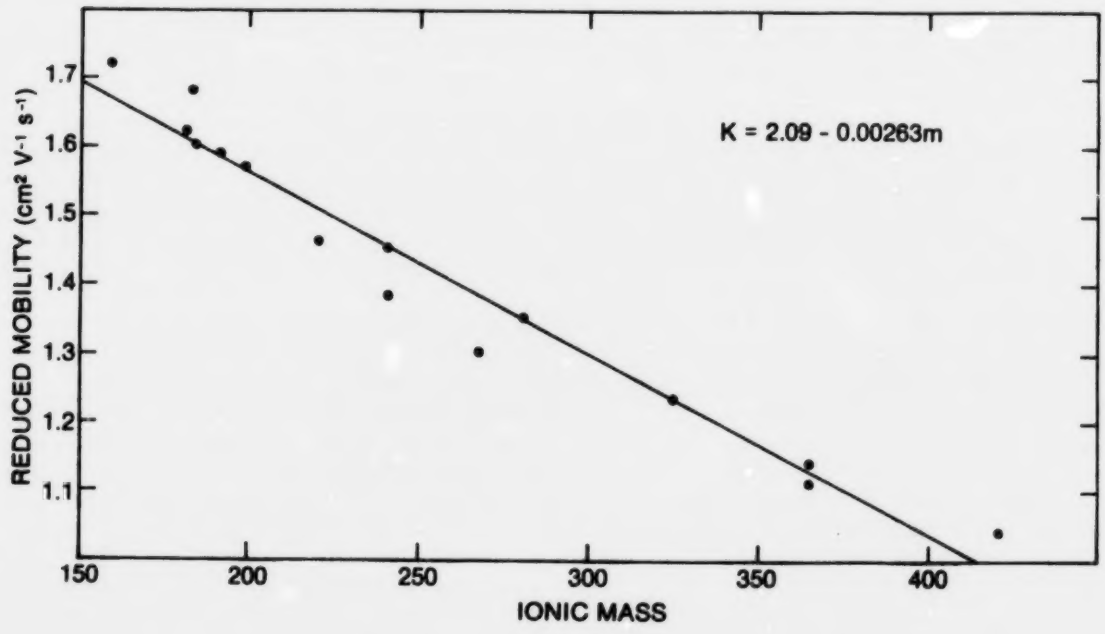

Figure 3. The reduced mobility of agent and simulant ions plotted against their masses on a linear scale.

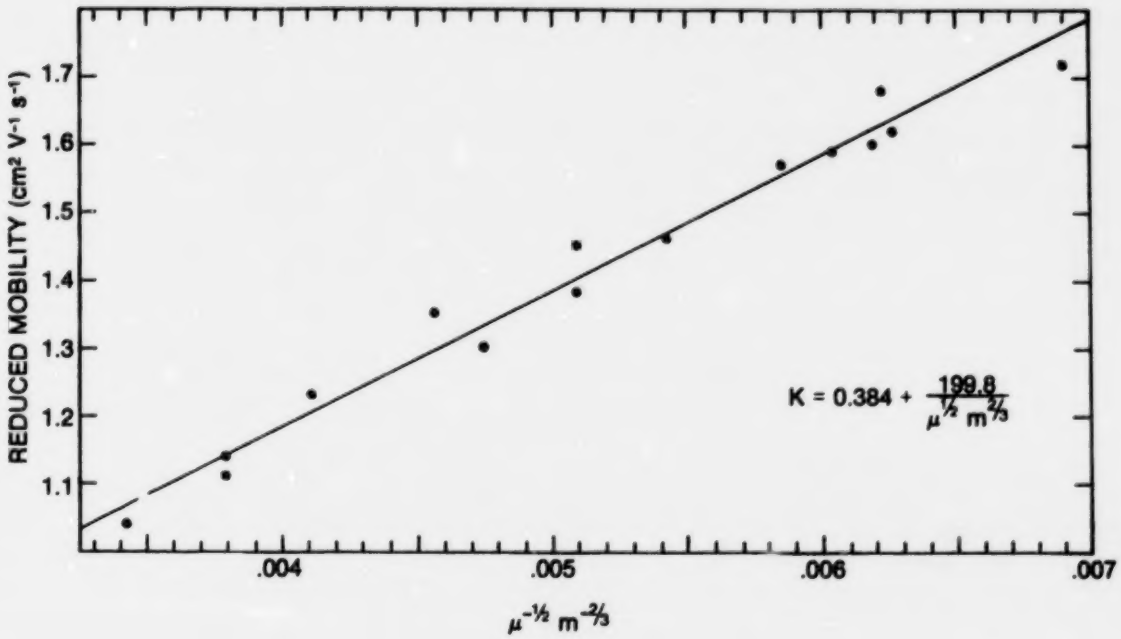

Figure 4. Another possible presentation of mobility-mass data is to plot mobility against this function, as described in the text.

other plots, for example in the graph of mobility plotted directly with ion mass (Figure 3).

As an intermediate between simplistic relationship such as linear and logarithmic plots, and the missing theory, there lies the "billiard ball theory" assumption of constant density of ionic matter. Here it is argued that the cross-section is simply the area presented by the ion to the gas molecules and therefore proportional to $m^{2/3}$ (m is the ion mass, ion assumed spherical). Equation 10 then gives

$$K \propto \mu^{-\frac{1}{2}} m^{-\frac{2}{3}} \qquad (21)$$

Figure 3 is a graph of mobility vs. $\mu^{-\frac{1}{2}} m^{-\frac{2}{3}}$. (The occurence of a non-zero intercept is hardly surprising since this crude model will fail in the limits of large or small ion mass.)

All three of these assumed relationships between ion mass and ion mobility seem to give reasonable plots. Is there anything to choose between them? One way of comparing the goodness of fit of the straight lines which have been fitted to these data by the least-squares method is to compute the standard error of estimate. This error is the root-mean-square sum of the differences between the experimental mobilities and the

mobility values predicted by the least-squares expression for that mass. For the three plots presented here these values are 0.044, 0.029, and 0.028 $cm^2 V^{-1} s^{-1}$ for the linear, logarithmic, and μ^{-1} $m^{-2/3}$ plots respectively. Our conclusion is that in the absence of appropriate theory several assumed relationships could give approximately straight-line plots, but that these correlations are not neccesarily significant. It seems that the justification for presenting mobility vs. mass data on a semilog plot is historical only, but is nevertheless convenient in the absence of adequate theory.

CONCLUSIONS

An examination of the theory of ion mobility has indicated that the complexity of the collisions prevents accurate calculations of the mobilities of agent ions in air. Calculations performed with the best and most complete theories available today are likely to be significantly in error for agent ions.

Development work which requires mobility data for these ions must rely on experimental values. Several plots for presenting this data in an organized format have been devised - they show good correlation but since they have no theoretical basis they could not be used to, for example, predict the mobility of a new agent ion.

The common practice of reducing mobility data to a standard temperature is not applicable to agent ions since the details of the collisions can change with temperature.

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It is a pleasure to acknowledge discussions with and help from D.A. Blyth, Chemical Defence Establishment, Porton Down, Wiltshire, England. He is the United Kingdom delegate to various TTCP Subgroup E activities which have been instrumental in advancing the development of ionization detectors in all countries participating. Mr. Blyth supplied the mobility data (Ref. 12), discussed the validity of reduced mobility and other aspects, and reviewed a draft of this note.

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